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STABLE O/W/O EMULSION AND ITS USE AS COSMETIC AND/OR
DERMATOLOGICAL COMPOSITION

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[Abstract]

The present invention relates to a triple oil/water/oil emulsion comprising a primary oil-in-water emulsion and an external oil phase, characterized in that it contains at least one

partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain, which is preferably introduced into the external oil phase.

The triple emulsion remains stable and it is particularly appropriate as a composition for topical administration, notably cosmetic or dermatological, in particular as a vehicle of active ingredients, in particular liposoluble active ingredients present in the internal oil phase.

The emulsion obtained can, in particular, constitute a composition to clean and/or treat and/or protect the skin and/or the mucosal membranes and/or the keratin fibers.

Description

[0001]

The present invention relates to a stable triple oil/water/oil emulsion and its use, notably in the cosmetic and/or dermatological fields, in particular for the controlled release of active ingredients, notably in view of cleaning, treating, protecting and/or hydrating the skin and/or the mucosal membranes and/or the keratin fibers and, more particularly, in view of treating dry skin.

[0002]

It is known to use, notably in the cosmetic and dermatological fields, topical compositions in the form of emulsions. These emulsions generally are oil-in-water (O/W) or water-in-oil (W/O) emulsions. They can also be multiple emulsions of the water/oil/water (W/O/W) or oil/water/oil (O/W/O) type. Among the multiple emulsions, it is preferred to use those with an external aqueous phase, namely W/O/W emulsions, which combine the advantages of freshness upon application, contributed by the water present in the aqueous external phase, and of the comfort contributed by a relatively large quantity of oil.

[0003]

However, the O/W/O emulsions also present the advantage, due to their oily continuous phase, of allowing the formation of a lipid film on the surface of the skin which prevents the transepidermal loss of water and protects the skin against external stress. In addition, one can prevent the fatty effect of such emulsions by incorporating in them new light oils, which are not greasy to the touch, such as, for example, silicone oils with a low viscosity, certain esters of fatty acids and short chain fatty alcohols. Therefore, it is of interest to have available multiple emulsions of the O/W/O type.

[0004]

However, the multiple emulsions are in general not used much, because they frequently present stability problems over time. The most frequently encountered destabilization mechanism is the migration of oil from internal droplets toward the external oily medium through the intermediate aqueous layer, either by a simple diffusion of oil through the aqueous membrane, or by prior rupture of the aqueous film causing the coalescence of internal drops of oil and leading to the release of oil into the external oily medium. In general, this phenomenon, which is referred to as loss of multiple character, ends up leading to a macroscopically visible dephasing and to the obtention of an unstable simple O/W emulsion instead of a triple emulsion.

[0005]

Different means have been considered to overcome this drawback. In particular, one of the solutions consists in introducing into the internal or external oily phase one or more gelling polymers whose role is to permanently limit the movements of oil from the internal phase toward the external phase. However, the polymers which are capable of gelling the oils are not very widespread and they do not have good cosmetic properties: they accentuate the greasy sensation and they are sticky during and after application to the skin. The multiple emulsions obtained present the defect of being sticky and of taking a long time to penetrate into the skin.

[0006]

Therefore, there remains a need for a stable O/W/O multiple emulsion which does not have the drawbacks of those of the prior art and which is, notably, pleasant to use on the skin.

[0007]

The applicant unexpectedly found that the use of a partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain allowed the obtention of a stable multiple O/W/O emulsion without requiring the addition of other stabilizing agents.

[0008]

Moreover, the present invention also relates to a triple oil/water/oil emulsion comprising a primary oil-in-water emulsion and an external oily phase, characterized in that the triple emulsion contains at least one partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain.

[0009]

The present invention also relates to the use of at least one partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain for the stabilization of a triple oil/water/oil emulsion.

[0010]

The triple emulsion according to the invention has the advantage of being stable and, notably, of being able to preserve the activity of active ingredients, notably lipophilic active ingredients, present in the internal oily phase, from which they are released during the application of the composition to the skin, the mucosal membranes and/or the hair.

[0011]

The partially or completely crosslinked organopolysiloxane elastomers which can be used in the emulsion according to the present invention can be introduced into either one of the oily phases. They are preferably introduced into the external oily phase of the emulsion. In general they are emulsifiers. They can notably be chosen from the crosslinked polymers described in the application EP-A-0545002 which is indicated here by reference. These organopolysiloxanes are obtained by the addition polymerization of the following compounds (I) and (II):

(I) an organohydrogen polysiloxane having formula (1):



in which R^1 represents a substituted or unsubstituted alkyl, aryl or aralkyl group comprising 1-18 carbon atoms or a halogenated hydrocarbon group; R^2 represents a group:



in which R^3 is a hydrogen, a saturated aliphatic hydrocarbon group having 1-10 carbon atoms or a group $-(CO)-R^5$ where R^5 is a saturated aliphatic hydrocarbon group having 1-5 carbon atoms; d is a whole number ranging from 2 to 200, and e is a whole number ranging from 0 to 200, provided that d + e is a whole number ranging from 3 to 200, and n is a number ranging from 2 to 6, a is a value satisfying the inequality: $1.0 \leq a \leq 2.5$, b is a value satisfying the inequality: $0.001 \leq b \leq 1.0$ and c is a value satisfying the inequality: $0.001 \leq c \leq 1.0$;

or an organohydrogen polysiloxane represented by the following formula (2):

* [Editor's note: the representations of the R groups in the original document is inconsistent.]



in which R^1 has the same meaning as in formula (1), f is a value satisfying the inequality:

$1.0 \leq f \leq 3.0$, g is a value satisfying the inequality: $0.001 \leq g \leq 1.5$;

or a mixture of organohydrogen polysiloxanes having formulas (1) and (2), and

(II) a polyoxyalkylene represented by the following formula (A):



in which h is a whole number ranging from 2 to 200, i is a whole number ranging from 0 to 200,

provided that $h + i$ is a number ranging from 3 to 200, and m is a number ranging from 2 to 6,

or an organopolysiloxane represented by the following formula (B):



in which R^1 has the same meaning as in formula (1), R^4 is a monovalent hydrocarbon group

having a saturated aliphatic bond at the end and containing 2-10 carbon atoms, j is a value

satisfying the inequality: $1.0 \leq j \leq 3.0$ and k is a value satisfying the inequality: $0.001 \leq k \leq 1.5$,

or a mixture of the polyoxyalkylene having formula (A) and the organopolysiloxane having

formula (B), where at least one organohydrogen polysiloxane having formula (1) or at least one

polyoxyalkylene having formula (A) is contained as an essential element of the addition

polymerization.

[0012]

Preferably, the organopolysiloxane is in a mixture with a silicone oil and/or a polyol and is directly prepared in such a mixture. The silicone oil presents, preferably, a viscosity which is equal to or less than 100 cSt at 25°C. According to an embodiment variant of the invention, the organopolysiloxane elastomer is prepared from 100 parts by weight of the constituents defined above and 3-200 parts by weight of a silicone oil having a viscosity equal to or greater than 100 cSt at 25°C, and/or a polyol. The silicone oil can be a volatile or nonvolatile silicone oil or a mixture of a volatile silicone oil and a nonvolatile silicone oil.

[0013]

As an example of a partially or completely crosslinked organopolysiloxane comprising a polyoxyethylenated and/or polyoxypropylenated chain one can mention the product marketed by Shin-Etsu under the name KSG 21. This product comprises 28% organopolysiloxane and 62% silicone oil having a viscosity of 6 cSt.

[0014]

In the triple emulsion according to the invention, the partially or completely crosslinked organopolysiloxane is preferably used in a quantity of active ingredient of 0.1-10 wt%, preferably 1-5 wt% with respect to the total weight of the triple emulsion.

[0015]

It is preferred for the primary O/W emulsion to contain one or more emulsifiers chosen from:

(1) nonionic surfactants having an HLB greater than or equal to 11, optionally combined with a lipophilic cosurfactant, such as a fatty alcohol, a fatty acid or a glyceryl fatty ester (glyceryl stearate). Examples of nonionic surfactants having an HLB greater than or equal to 11 include oxyethylenated fatty acid and glycerol esters, oxyethylenated fatty acid and sorbitan esters, oxyethylenated fatty acids, sugar esters such as sucrose stearate, and their mixtures;

(2) polymers capable of stabilizing an O/W emulsion. Examples of polymers of this type include copolymers consisting of a majority fraction of a monoolefinically unsaturated C₃-C₆ carboxylic acid monomer or its anhydride and a minority fraction of an acrylic acid fatty ester monomer, such as the product marketed under the name Pemulen TR2 by the Goodrich Company (CTFA name: acrylates/C10-30 alkyl acrylate cross polymer), or polyacrylamides, such as the product marketed under the name Hostacerin AMPS by the Hoechst Company (CTFA name: ammonium polyacryldimethyl tauramide), and their mixtures;

(3) dispersions of lipid vesicles based on ionic amphiphilic lipids (liposomes), or nonionic amphiphilic lipids, and notably liposomes based on the combination of hydrogenated lecithin/oxyethylenated soy sterol.

[0016]

One can also use a mixture of these emulsifiers.

[0017]

These emulsifiers can be introduced into the aqueous phase or into the oily phase of the primary emulsion.

[0018]

The quantity of emulsifier(s) in the primary emulsion varies depending on the nature of the emulsifiers used. It can range, for example, from 0.1 to 15 wt% with respect to the total weight of the primary emulsion.

[0019]

The quantity of primary O/W emulsion in the triple emulsion is generally 50-95 wt%, preferably 70-85 wt% with respect to the total weight of the triple emulsion.

[0020]

The quantity of the internal oily phase is generally 0.1-40 wt%, preferably 1-25 wt% with respect to the total weight of the triple emulsion. The quantity of the aqueous phase is generally 10-90 wt%, preferably 40-80 wt% with respect to the total weight of the triple emulsion.

[0021]

The oily phase of the O/W primary emulsion and the external oily phase comprise one or more fatty substances chosen from oils of animal origin, oils of plant origin (apricot almond oil, liquid fraction of shea butter), mineral oils (vaseline), synthetic oils (isohexadecane, hydrogenated polyisobutene or parleam oil), fluorinated oils, silicone oils and notably volatile silicone oils, such as octyl heptamethyltrisiloxane (or caprylylmethicone) and cyclomethicones such as cyclopentasiloxane and cyclohexasiloxane, waxes, notably silicone waxes, silicone gums, and silicone resins.

[0022]

It is preferred for the oily phase, which contains the organopolysiloxane elastomer and notably the external oily phase of the multiple O/W/O emulsion, to contain, as a fatty substance, at least one solvent of the organopolysiloxane elastomer, which solvent is preferably a silicone oil chosen from cyclic organopolysiloxanes (cyclomethicone), such as cyclohexamethylsiloxane or low-viscosity linear organopolysiloxanes (polydimethylsiloxanes having a viscosity of less than 50 cSt or dimethicones).

[0023]

The triple emulsion is prepared in the conventional manner by the preparation of the primary emulsion and incorporation of a predetermined quantity of the primary emulsion in the external oily phase which, according to a preferred embodiment variant of the invention, contains the partially or completely crosslinked organopolysiloxane elastomer which comprises a polyoxyethylenated and/or polyoxypropylenated chain.

[0024]

The triple emulsion according to the invention can notably be used in the cosmetic, dermatological and pharmaceutical fields. Preferably, it is intended to constitute a composition

for topical administration. In this case, the composition must contain a topically acceptable medium, that is one which is compatible with the skin, the mucosal membranes, the nails, the scalp and/or the hair. As indicated at the beginning of the description, one of the major advantages of the emulsion according to the invention is that the latter can contain, while presenting a stable character, both cosmetic and therapeutic active ingredients, where these active ingredients can thus notably be chosen from all those that are conventionally used to date in the field of cosmetics, dermatology or drugs.

[0025]

This active ingredient can notably be a lipophilic active ingredient, but it can also be hydrophilic, and, depending on its nature, it can be introduced into one of the oily phases or into the aqueous phase of the triple emulsion according to the invention.

[0026]

Lipophilic active ingredients in particular include lipophilic vitamins such as vitamin A (retinol), vitamin D, vitamin E (tocopherol), vitamin K and derivatives of these vitamins, such as esters, ceramides, unsaponifiable components such as unsaponifiable component of shea butter algal extracts, notably those which are rich in polyunsaturated fatty acids, such as eicosapentaenoic acid and docosahexaenoic acid, unsaturated oils, for example, fish oils which are rich in linoleic and linolenic acids.

[0027]

As hydrophilic active ingredients, one can notably cite polyols, such as glycerin, glycols and sugar derivatives, enzymes, natural extracts, procyanidolic oligomers, vitamins, such as vitamin C and its derivatives, as well as esters, urea, depigmentation agents, as well as kojic acid and caffeic acid, beta-hydroxy acids, such as salicylic acid and its derivatives, alpha-hydroxy acids such as lactic acid and glycolic acid, hydrating agents such as protein hydrolysates, and softeners, such as allantoin and their mixtures.

[0028]

The active ingredient is present in an effective quantity to ensure the expected result. For example, it can be present in the emulsion according to the invention in a quantity of 0.01-20 wt%, preferably 0.1-10 wt%, and more advantageously 0.5-5 wt% with respect to the total weight of the composition.

[0029]

The O/W/O emulsions according to the invention can notably constitute cleaning, protection, treatment and/or care compositions for the skin, the mucosal membranes and/or the hair, in particular for the face, neck, hands, hair, scalp or body, as well as eyelashes. They can notably constitute protection, treatment or care creams for the face, hands, or feet, body milks for protection or care, lotions, gels or foams for the care of the skin, the mucosal membranes, the hair and/or scalp. The compositions of the invention are particularly appropriate for hydrating the skin and/or treating dry skin.

[0030]

Moreover, the invention also relates to the cosmetic use of the emulsion as defined above to clean and/or treat and/or protect and/or hydrate the skin and/or the mucosal membranes and/or the keratin fibers, that is the hair and/or eyelashes.

[0031]

The invention also relates to the use of the emulsion as defined above for the preparation of a dermatological composition intended for treating and/or protecting dry skin.

[0032]

The invention also relates to a cosmetic and/or dermatological method to clean and/or treat and/or protect and/or hydrate the skin, the mucosal membranes and/or the keratin fibers, characterized in that it consists in applying to the skin, the mucosal membranes and/or the keratin fibers an emulsion as defined above.

[0033]

In a known manner, the composition of the invention can also contain lipophilic or hydrophilic adjuvants which are conventional in the cosmetic and/or dermatological fields, such as foaming surfactants, preservatives, antioxidants, sequestering agents, solvents (octyldodecanol), perfumes, fillers, sunscreens, odor absorbents, dyes, the gelling agents and lipid vesicles.

[0034]

As gelling agents, one can cite, for example, clays, polysaccharide gums and their derivatives (xanthan gum, carboxymethylhydroxypropyl guar), and carboxyvinyl polymers (carbomer or sodium carbomer).

[0035]

The quantities of the different adjuvants are those conventionally used in the fields considered and, for example, 0.01-15 wt% of the total weight of the composition. The nature of the adjuvants and their quantities must be such that they do not modify the properties of the compositions according to the invention.

[0036]

The adjuvants, depending on their type, can be introduced into the aqueous phase or into one of the oily phases of the triple emulsion.

[0037]

The examples below of compositions according to the invention are given for illustration and without limiting character. The quantities given therein are in wt% with respect to the total weight of the composition.

Example 1: Hydrating night cream

1. Primary O/W emulsion:

Phase A: Preparation of liposomes

[0038]

- Hydrogenated lecithin (Lecinol S 10 marketed by NIKKOL)	0.72%
- Oxyethylenated soy sterols (Generol 122 N E 5D marketed by HENKEL)	2.8%
- Water	9%

Phase B: Oil phase of the primary O/W emulsion

[0039]

- Isohexadecane	3.5%
- Apricot almond oil	5%
- Unsaponifiable components of shea butter	1%
- Perfume	0.5%
- Preservative	0.6%

Phase C: Aqueous phase

[0040]

- Glycerin	3%
- Preservative	0.2%
- Xanthan gum	0.2%

- Sodium carbomer	0.15%
- Water	53.33%

2. External oil phase:

[0041]

- Cyclohexasiloxane	12%
- KSG 21-28% of active ingredient	8%

Example 2: Nourishing creams for dry skins

1. Primary O/W emulsion:

Phase A:

[0042]

- Glyceryl stearate and PEG-100 glyceryl stearate (Arlacel 165 marketed by ICI)	1.8%
- Polysorbate 60 (Tween 60 marketed by ICI)	0.75%
- Liquid fraction of shea butter	7.3%
- Cetyl alcohol	1.75%
- Cyclopentasiloxane (DC Fluid 245 marketed by Dow Corning)	7%
- Parleam oil	4%
- Perfume	0.5%
- Preservative	0.6%

Phase D

- Glycerin	7%
- Preservative	0.2%
- Xanthan gum	0.1%
- Sodium carbomer	0.2%
- Water	53.8%

2. External oily phase:

[0044]

- Dimethicone 10 cSt (DC Fluid 200 marketed by Dow Corning)	10%
- KSG 21-28% of active ingredient	8%

Example 3: Hydrating cream

1. Primary O/W emulsion

Phase A:

[0045]

- Octyldodecanol	2%
- Cyclopentasiloxane (DC Fluid 245 marketed by Dow Corning)	4.5%
- Tocopherol acetate	1%
- Perfume	0.5%
- Preservative	0.6%

Phase B:

[0046]

- Glycerin	5.5%
- Preservative	0.2%
- Acrylate/C 10-30 alkyl acrylate cross polymer (Pemulen TR 2 marketed by GOODRICH)	0.13%
- Ammonium polyacryldimethyl tauramide (Hostacerin AMPS marketed by HOECHST)	1.1%
- Water	69.47%

2. External oily phase:

[0047]

- Caprylmethicone (Silsoft 034 marketed by WITCO)	7%
- KSG 21-28% of active ingredient	8%

Claims

1. Triple oil/water/oil emulsion comprising a primary oil-in-water emulsion and an external oil phase, characterized in that the triple emulsion contains at least one partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain.

2. Emulsion according to Claim 1, characterized by the fact that the organopolysiloxane elastomer is obtained by addition polymerization of the following compounds (I) and (II):

(I) an organohydrogen polysiloxane having formula (1):



in which R^1 represents a substituted or unsubstituted alkyl, aryl or aralkyl group comprising 1-18 carbon atoms or a halogenated hydrocarbon group; R^2 represents a group:



in which R^3 is a hydrogen, a saturated aliphatic hydrocarbon group having 1-10 carbon atoms or a group $-(CO)-R^5$ where R^5 is a saturated aliphatic hydrocarbon group having 1-5 carbon atoms; d is a whole number ranging from 2 to 200, and e is a whole number ranging from 0 to 200, provided that d + e is a whole number ranging from 3 to 200, and n is a number ranging from 2 to 6, a is a value satisfying the inequality: $1.0 \leq a \leq 2.5$, b is a value satisfying the inequality: $0.001 \leq b \leq 1.0$ and c is a value satisfying the inequality: $0.001 \leq c \leq 1.0$;

or an organohydrogen polysiloxane represented by the following formula (2):



in which R^1 has the same meaning as in formula (1), f is a value satisfying the inequality: $1.0 \leq f \leq 3.0$, g is a value satisfying the inequality: $0.001 \leq g \leq 1.5$;

or a mixture of organohydrogen polysiloxanes having formulas (1) and (2), and

(II) a polyoxyalkylene represented by the following formula (A):



in which h is a whole number ranging from 2 to 200, i is a whole number ranging from 0 to 200, provided that h + i is a number ranging from 3 to 200, and m is a number ranging from 2 to 6,

or an organopolysiloxane represented by the following formula (B):



which R^1 has the same meaning as in formula (1), R^4 is a monovalent hydrocarbon group having a saturated aliphatic bond at the end and containing 2-10 carbon atoms, j is a value satisfying the inequality: $1.0 \leq j \leq 3.0$ and k is a value satisfying the inequality: $0.001 \leq k \leq 1.5$, or a mixture of the polyoxyalkylene having formula (A) and of the organopolysiloxane having formula (B), where at least one organohydrogen polysiloxane having formula (1) or at least one polyoxyalkylene having formula (A) is contained as an essential element of the addition polymerization.

3. Emulsion according to Claim 1 or 2, characterized by the fact that the organopolysiloxane elastomer is present in the external oily phase.

4. Emulsion according to any one of the preceding claims, characterized by the fact that the organopolysiloxane elastomer is in a mixture with a silicone oil and/or a polyol.

5. Emulsion according to the preceding claim, characterized by the fact that the silicone oil preferably has a viscosity equal to or less than 100 cSt at 25°C.

6. Emulsion according to Claim 4 or 5, characterized by the fact that the silicone oil has a viscosity equal to 6 cSt at 25°C.

7. Emulsion according to any one of the preceding claims, characterized in that the organopolysiloxane elastomer is present in a quantity of active ingredient of 0.1-10 wt% with respect to the total weight of the triple emulsion.

8. Emulsion according to any one of the preceding claims, characterized in that the primary O/W emulsion contains one or more emulsifiers chosen from nonionic surfactants having an HLB greater than or equal to 11, polymers which are capable of stabilizing an O/W emulsion, dispersions of lipid vesicles and their mixtures.

9. Emulsion according to the preceding claim, characterized in that the nonionic surfactant is chosen from oxyethylenated esters of fatty acid and glycerol esters, oxyethylenated fatty acid and sorbitan esters, oxyethylenated fatty acids, sugar esters, and their mixtures.

10. Emulsion according to Claim 8, characterized in that the polymer is chosen from copolymers consisting of a majority fraction of a monoolefinically unsaturated C₃-C₆ carboxylic acid monomer or its anhydride and a minority fraction of an acrylic acid fatty ester monomer, polyacrylamides, and their mixtures.

11. Emulsion according to Claim 8, characterized in that the dispersion of lipid vesicles is a dispersion of liposomes.

12. Emulsion according to any one of the preceding claims, characterized in that the oily phases contain one or more fatty substances chosen from oils of animal origin, oils of plant origin, mineral oils, synthetic oils, silicone oils, fluorinated oils, waxes, silicone gums, and silicone resins.

13. Emulsion according to any one of the preceding claims, characterized in that the quantity of internal oily phase is 0.1-40 wt%, preferably 1-25 wt% with respect to the total weight of the triple emulsion.

14. Emulsion according to any one of the preceding claims, characterized in that the quantity of primary O/W emulsion is 50-95 wt%, preferably 70-85 wt% with respect to the total weight of the triple emulsion.

15. Emulsion according to any one of the preceding claims, characterized in that it constitutes a composition for topical administration.

16. Emulsion according to any one of the preceding claims, characterized in that it contains at least one active ingredient chosen from vitamins, ceramides, unsaponifiable components, algal extracts, unsaturated oils, polyols, enzymes, natural extracts, procyanidolic

oligomers, urea, depigmentation agents, beta-hydroxy acids, alpha-hydroxy acids, hydrating agents, softeners, and their mixtures.

17. Emulsion according to the preceding claim, characterized in that the quantity of active ingredient(s) is 0.01-20 wt%, preferably 0.1-10 wt% with respect to the total weight of the triple emulsion.

18. Emulsion according to any one of the preceding claims, characterized in that it comprises at least one lipophilic or hydrophilic adjuvant chosen from preservatives, antioxidants, sequestering agents, solvents, perfumes, fillers, sunscreens, odor absorbents, dyes, gelling agents and lipid vesicles.

19. Cosmetic use of the emulsion according to any one of the preceding claims to clean and/or treat and/or protect and/or hydrate the skin, the mucosal membranes and/or the keratin fibers.

20. Use of the emulsion according to any one of Claims 1-18 for the preparation of a dermatological composition intended for treating and/or protecting dry skin.

21. Cosmetic method for cleaning and/or treating and/or protecting and/or hydrating the skin and/or the keratin fibers, characterized in that it consists in applying to the skin, the mucosal membranes and/or the keratin fibers an emulsion according to any one of Claims 1-18.

22. Use of at least one partially or completely crosslinked organopolysiloxane elastomer comprising a polyoxyethylenated and/or polyoxypropylenated chain, for the stabilization of a triple oil/water/oil emulsion.

European
Patent Office
EUROPEAN SEARCH REPORT

Application Number
EP 99 40 1883

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int Cl ⁷)
X	EP 0 559 013 A (TH.GOLDSCHMIDT AG) September 8, 1993 (1993-09-08) * Claims 1-5 * -----	1-4	A61K7/00 A61K7/48 B01F17/00
			TECHNICAL FIELDS SEARCHED (Int. Cl. ⁷)
			A61K B01F
The present search report has been drawn up for all claims.			
Place of search THE HAGUE		Date of completion of the search December 15, 1999	Examiner Fouquier, J-P
CATEGORY OF CITED DOCUMENTS			
X: Particularly relevant if taken alone.		T: Theory or principle underlying the invention.	
Y: Particularly relevant if combined with another document of the same category.		E: Earlier patent document, but published on, or after the filing date.	
A: Technological background.		D: Document cited in the application.	
O: Non-written disclosure.		L: Document cited for other reasons.	
P: Intermediate document.		&: Member of the same patent family, corresponding document.	

APPENDIX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN
PATENT APPLICATION NO.

EP 99 40 1883

In this appendix, the patent family members of patent documents listed in the above-referenced European Search Report are indicated.

The data on the family members correspond to the state of the files of the European Patent Office on December 15, 1999
These data serve only for information and are given without guarantee by the European Patent Office.

Patent document listed in the search report	Date of publication	Member(s) of the patent family	Date of publication
EP 559013 A	September 8, 1993	DE 4206732 A	September 9, 1993
		DE 59300435 D	September 14, 1995
		ES 2076799 T	November 1, 1995
		JP 5337359 A	December 21, 1993
		US 5391321 A	February 21, 1995